

We Claim :

1. A process for the synthesis of a α -substituted acrolein of the formula $\text{CH}_2=\text{CR}-\text{CHO}$ where R is an alkyl or aryl group, the process comprising

i. subjecting a mixture of an olefin and syngas or a mixture of olefin and carbon monoxide and hydrogen to hydroformylation in organic phase and in the presence of a hydroformylation catalyst or catalyst precursor comprising a rhodium complex catalyst or catalyst precursor to obtain the corresponding aldehyde;

ii. subjecting the aldehyde obtained in step (i) above to aldol condensation with formaldehyde in the presence of an aldolisation catalyst comprising secondary amine or a secondary amine/organic acid catalyst in aqueous phase,

the reactions of steps (i) and (ii) being carried out simultaneously in a biphasic aqueous-organic system, ensuring the segregation of the two catalysts into two distinct phases, to obtain an α -substituted acrolein.

2. A process as claimed in claim 1 wherein the rhodium complex catalyst comprises an organometallic complex of rhodium of the formula $\text{Rh}(\text{A})_p(\text{B})_q(\text{C})_r(\text{D})_s$ wherein A, B, C and D are ligands selected from the group consisting of H, trialkyl phosphines, triaryl phosphines, carbonyl, trialkyl arsine, triaryl arsines, alkyl aryl phosphines, trialkyl amines, triaryl amines, alkyl aryl amines, bisphosphines and diimines and $p+q+r+s$ is equal to 4, 5 or 6, such that the resultant complex is soluble in the organic phase.

3. A process as claimed in claim 1 wherein the organic phase comprises an organic media selected from the group consisting of aromatic hydrocarbons, aliphatic hydrocarbons, higher alcohols and any mixtures thereof.

4. A process as claimed in claim 1 wherein the rhodium complex catalyst is selected from the group consisting of $\text{HRh}(\text{CO})(\text{PPh}_3)_3$, $\text{Rh}(\text{CO})(\text{Cl})(\text{PPh}_3)_2$, $\text{Rh}(\text{CO})_2(\text{acetylacetonate})$, $[\text{Rh}(\text{Cyclooctadiene})\text{Cl}]_2$, $\text{HRh}(\text{CO})(\text{Pn-butyl})_3$, $\text{HRh}(\text{CO})(\text{PPh}_3)(\text{diphenylphosphinoethane})$, $\text{HRh}(\text{CO})(\text{PPh}_3)(\text{diphenylphosphinopropane})$ and $\text{HRh}(\text{CO})(\text{PPh}_3)(\text{diphenylphosphinbutane})$.

5. A process as claimed in claim 1 wherein the step of hydroformylation is carried out using a rhodium catalyst precursor with an additional ligand and in the presence of an additional ligand which identical to or different from the ligand already coordinated to the rhodium metal precursor.

6. A process as claimed in claim 5 wherein the additional ligand is water insoluble and is selected from the group consisting of aryl, alkyl and alkylaryl secondary phosphines, aryl, alkyl and alkylaryl tertiary phosphines, aryl, alkyl and alkylaryl secondary phosphates,

aryl, alkyl and alkylaryl tertiary phosphates, aryl, alkyl and alkylaryl secondary arsines, aryl, alkyl and alkylaryl tertiary arsines, aryl, alkyl and alkylaryl tertiary amines, pentanediones, substituted pentanediones, acetylacetonates, Schiff bases and aryl, alkyl and alkylaryl bisphosphines.

- 5 7. A process as claimed in claim 5 wherein the additional ligand is selected from the group consisting of triphenylphosphine, tri n-butylphosphine, acetylacetone, tributylphospite, triphenylphosphite, triphenylamine, tributylamine and triphenylarsine.
8. A process as claimed in claim 5 wherein the reaction is carried out in presence of excess ligand and mole ratio of metal to free ligand is in the range of 0.1 to 10.
- 10 9. A process as claimed in claim 1 wherein the aldolisation catalyst is soluble in aqueous medium and comprises a secondary aryl or alkyl amine selected from the group consisting of diethyl amine, dimethyl amine, methyle ethyl amine, dibutly amine, dibenzyl diphenyl amine, piperidine, morpholine, piperazine and pyrrolidine,
10. A process as claimed in claim 1 wherein the aldolisation reaction is carried out in the
15 presence of an organic acid selected from acetic acid, propionic acid and butyric acid.
11. A process as claimed in claim 10 wherein the mole ratio of secondary amine aldolisation catalyst to the organic acid is in the range of 0.01-10.
12. A process as claimed in claim 1 wherein the mole ratio of hydroformylation catalyst to the aldolisation catalyst is in the range of 0.01 to 10000
- 20 13. A process as claimed in claim 1 wherein the mole ratio of hydroformylation catalyst to the aldolisation catalyst is in the range of 0.1 to 1000.
14. A process as claimed in claim 1 wherein the organic phase of the present invention is immiscible with water and is selected from the group consisting of toluene, xylene, cyclohexane, heptane, decanol and any mixture thereof.
- 25 15. A process as claimed in claim 1 wherein the formaldehyde is used in the form of an aqueous solution or in the form of paraformaldehyde.
16. A process as claimed in claim 1 wherein the mole ratio of olefin to formaldehyde is in the range of 0.1 to 100.
17. A process as claimed in claim 1 wherein the mole ratio of olefin to formaldehyde is in the
30 range of 0.5 to 10.
18. A process as claimed in claim 1 wherein the olefin is selected from substituted and unsubstituted olefins with a carbon number from 2 to 10.
19. A process as claimed in claim 1 wherein the reaction temperature varies between 20 to 200 °C.

20. A process as claimed in claim 1 wherein the pressure of carbon monoxide and hydrogen is in the range of 10 to 1500 psi each.
21. A process as claimed in claim 1 wherein the pressure of carbon monoxide and hydrogen is in the range of 50 to 450 psi each.
- 5 22. A process as claimed in claim 1 wherein the product α substituted acrolein is preferentially soluble in the organic media.
23. A process as claimed in claim 1 wherein the reaction is conducted either as a batch or a continuous reaction with continuous addition of olefin and carbon monoxide and hydrogen dependant on consumption rate thereof.
- 10 24. A process as claimed in claim 1 wherein the olefin used is ethylene and the product obtained is methacrolein.

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